Phase-Field Modeling of Corrosion for Design of Next-Generation Magnesium-Aluminum Vehicle Joints

Presenter:
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Project ID: mat151

Overview

Timeline

Start Date: October, 2018

End Date: June, 2022

Budget

Total Project Funding: $ 1,899,462

- DOE: $ 1,499,612
- Participants: $ 399,850 (21%)

Actual Costs Incurred: $ 671,160

- DOE: $ 509,213
- Participants: $ 161,947 (24%)

As of December 31, 2019

Status: 33% of time, 34% of DOE budget

Any proposed future work is subject to change based on funding levels

Barriers & Technical Targets

- Barrier: limited understanding of multi-material joint corrosion & fracture – Mg-Al, friction stir welds (FSW)
- Target: validated model of microgalvanic corrosion and mechanical failure based on joint microstructure
- **Accomplishments**
- Diffusion bonded pure Al-Mg and 6022-ZEK100
- Began corrosion testing of diffusion bonds
- Phase field diffusion bond model, corrosion formula
- Developed FSW method for pure and alloy Al-Mg

Technology Partners

Worcester Polytechnic University (WPI)

Pacific Northwest Laboratory (PNNL)

Oak Ridge National Laboratory (ORNL)

Magna International, Inc. (Magna)

Relevance and Project Objectives

Relevance DOE VTO Materials Team Roadmap Multi-Material Systems Enablers: high-volume joining, corrosion, predictive modeling

Objective Develop and validate phase field corrosion model and coupled mechanical failure model in magnesium-aluminum alloy joints

End-of-Project Goal Predict tensile & fatigue strength of corroded joints within 10% of measured values

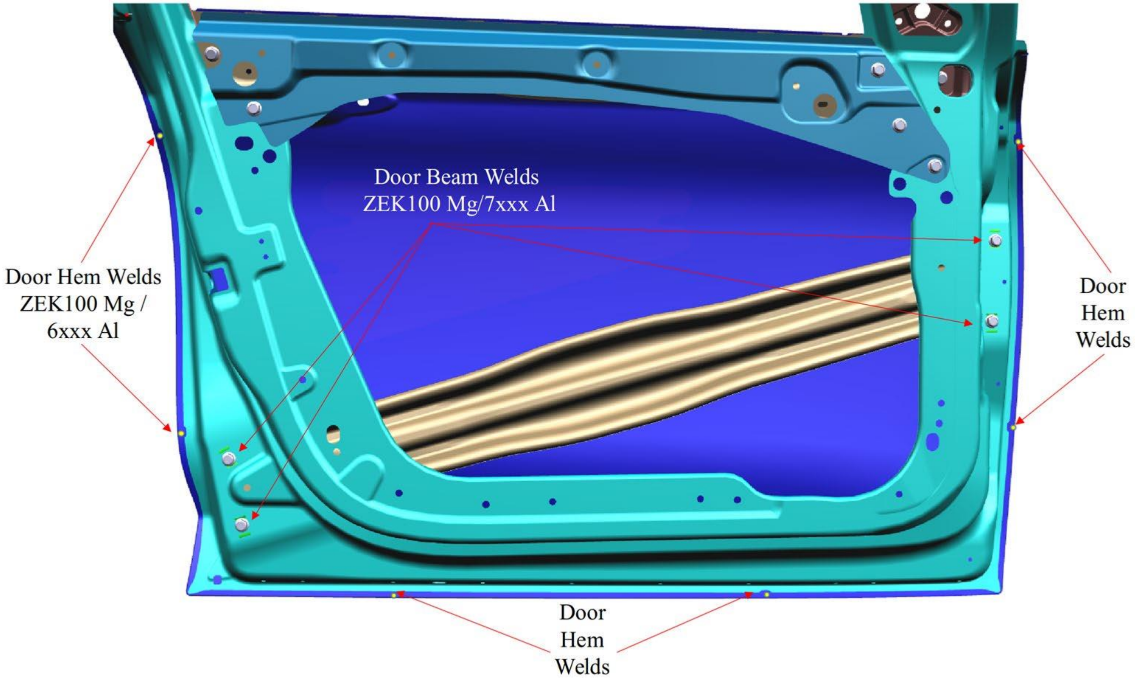
Magna – Joint application requirements, materials

PNNL – Friction stir welding, diffusion bonds (Task 1); modeling consulting (Task 4)

ORNL – Advanced characterization (Task 3)

WPI – Corrosion and mechanics testing (Task 2), corrosion and mechanics modeling (Task 4)

Challenge problem FCA-Magna ultralight door



Deliverables:

6/2020 Validated 2D galvanic corrosion model

6/2021 Validated model of coupled corrosion and mechanical failure

6/2022 Accurate model of corroded joint tensile & fatigue strength

Project Approach and Milestones

Time	Goal	Bond (PNNL)	Tests (WPI)	ORNL	Model (WPI)
BP 1: 10/2018-6/2020	Initial phase field corrosion model: diffusion bond	Diffusion-bond Al-Mg sheet, 6022-ZEK100	Galvanic corrosion - ASTM G71	SEM-EDS, EBSD, FIB, STEM	2-D diffusion bond & galvanic corrosion models
	Go/No Go	**Predict corrosion pit depth within ±2x**			
BP 2: 7/2020 - 6/2021	Refine corrosion & initial strength model	FSW 6022-ZEK100 sheet	Cyclic Corrosion Testing (CCT), tensile strength	SEM, STEM, FIB, neutron scattering	3-D corrosion & tensile failure models
	Go/No Go	**Predict corroded joint strength within ±2x**			
BP 3: 7/2021 - 6/2022	Refine tensile and fatigue strength models	FSW 6022-ZEK100 sheet	Cyclic Corrosion Testing (CCT), tensile strength	SEM, STEM, neutron scattering	3-D corrosion & tensile and fatigue models
	Go/No Go	**Corroded joint tensile & fatigue strength ±10%**			

S(T)EM: Scanning (Transmission) Electron Microscopy
EDS: Energy-Dispersive X-Ray Spectroscopy
EBSD: Electron Backscatter Diffraction
FIB: Focused Ion Beam (milling)

Project Accomplishments & Progress

Diffusion bonding aluminum to magnesium

- Established protocols and made samples
- Validated model of multi-phase interdiffusion

Corrosion of Al, Mg, alloys, diffusion bonded couples

- Measured corrosion rates of pure Al and Mg, 6022 and ZEK100
- Verified Cahn-Hilliard phase field model of galvanic reaction
- Designed 4-component Cahn-Hilliard formulation to describe multi-phase corrosion with water oxidation reaction at cathode

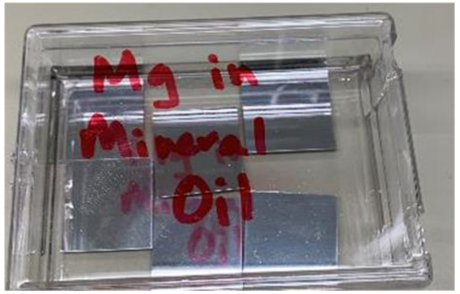
Friction stir welding through 6022 Al into ZEK100 Mg

- Conducted extensive parametric study of FSW through Al into Mg
- Established protocol with good repeatability and strength

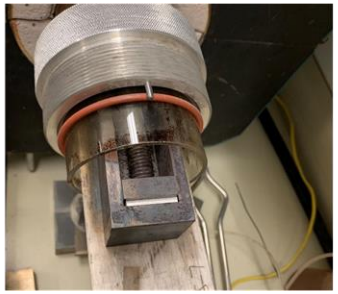
Diffusion Bonding: Protocols and Samples

Pacific Northwest NATIONAL LABORATORY

Diffusion Bonding Set-Up

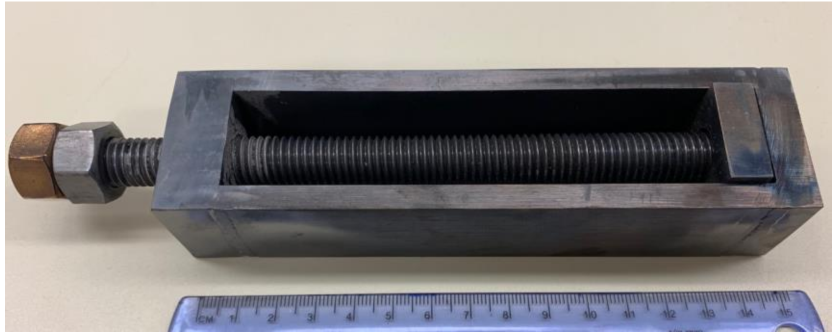


Mg in Oil



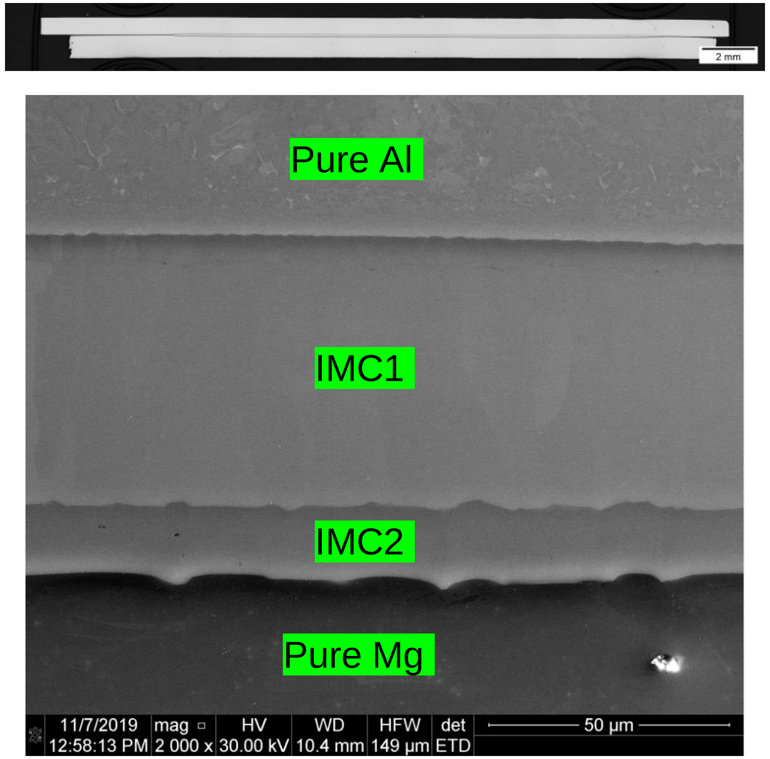
Side View

Sample Holder



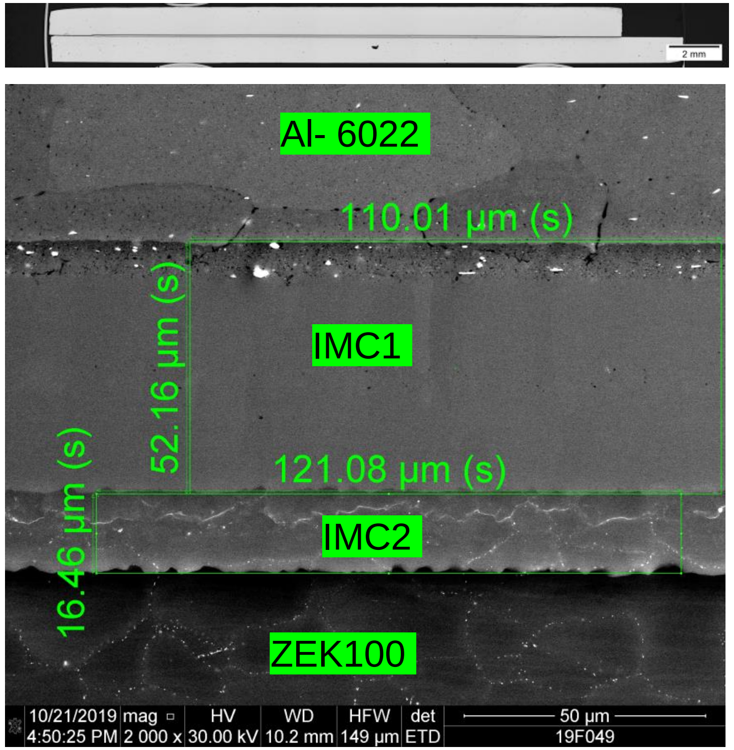
We established diffusion bonding protocols including sample preparation, storage, temperature and time for high quality diffusion bonding for corrosion testing and model development.

Pure Al- Pure Mg



395C for 4 hours yielded edge to edge diffusion bond in Pure Al-Pure Mg

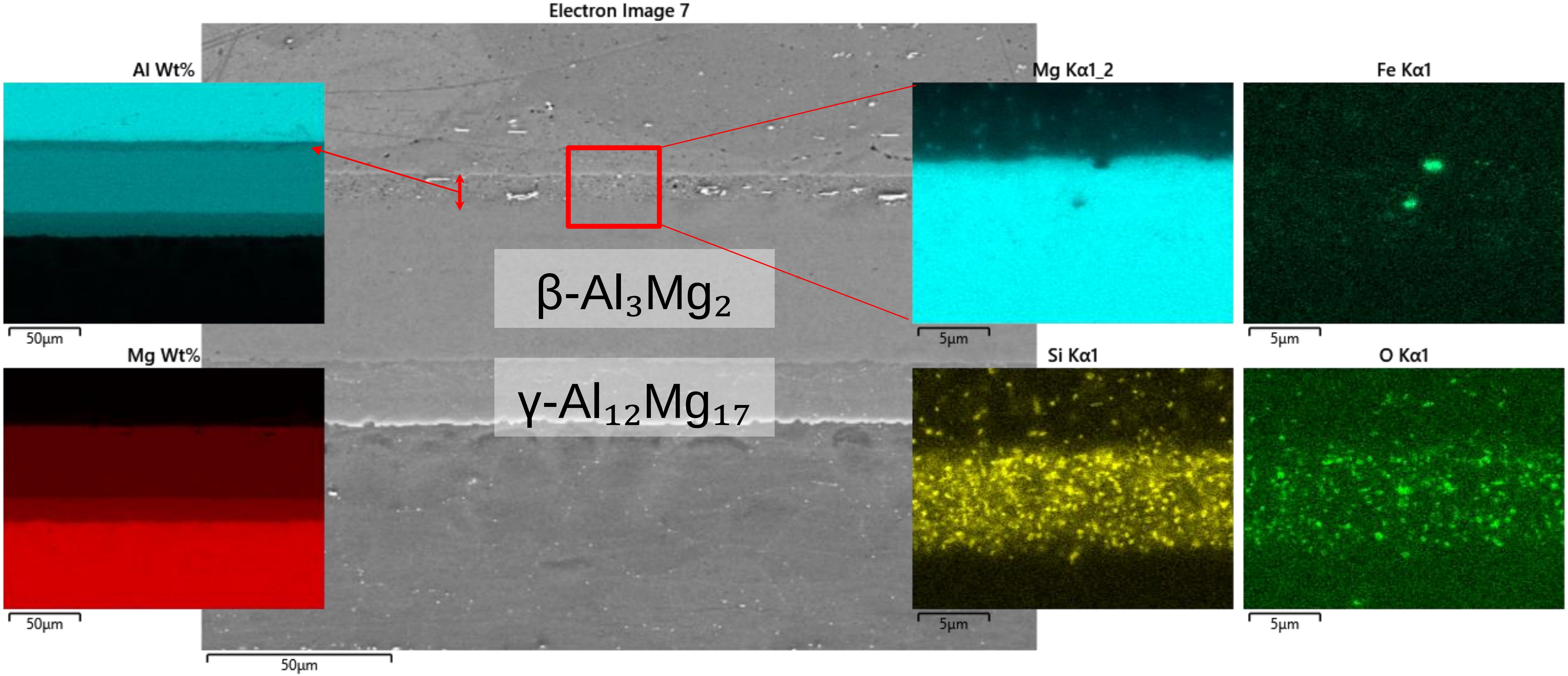
AA6022- ZEK100



395C for 2 hours yielded edge to edge diffusion bond in 6022- ZEK 100

Pure metals require more time for a good bond

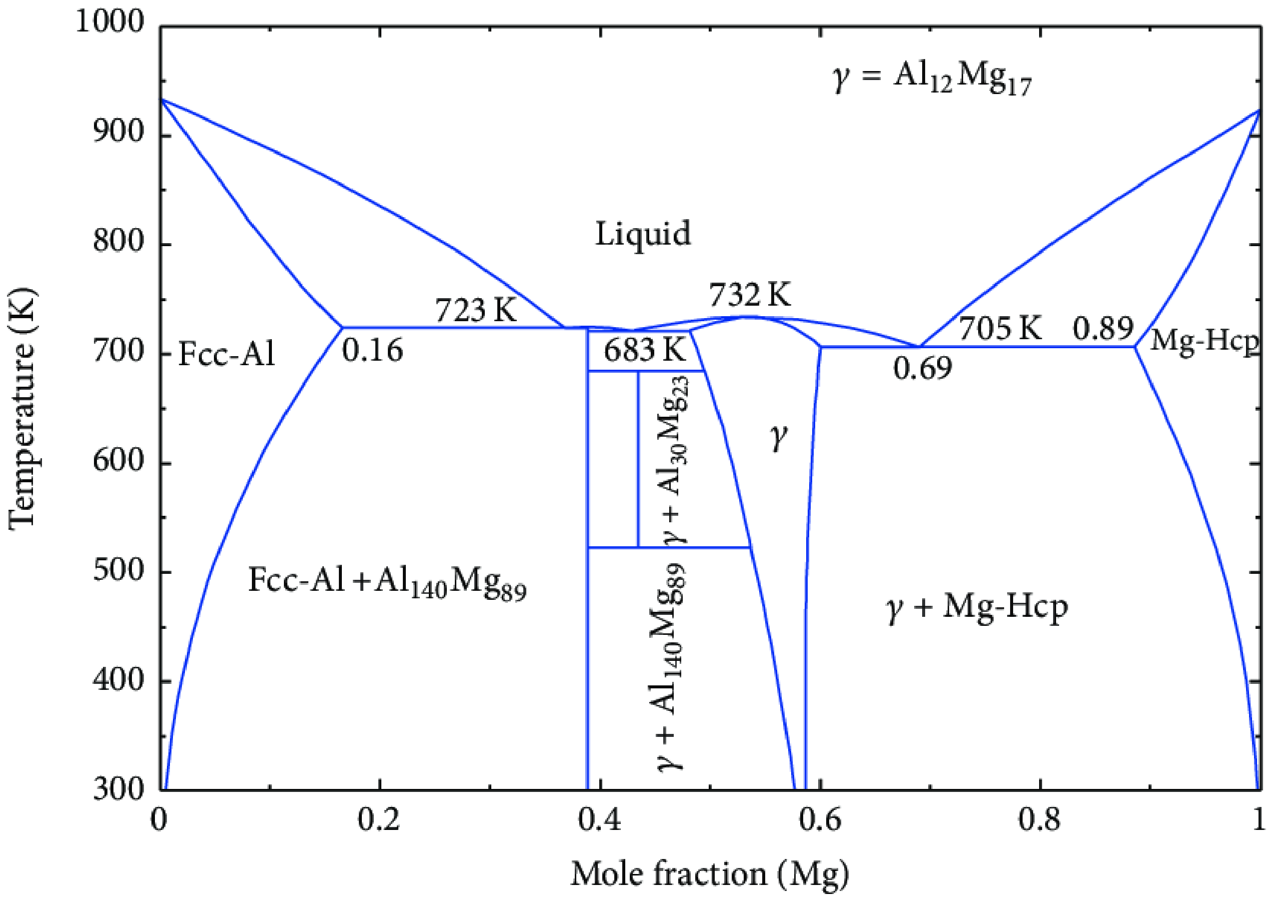
Diffusion Bonding: Nanoscale Microanalysis

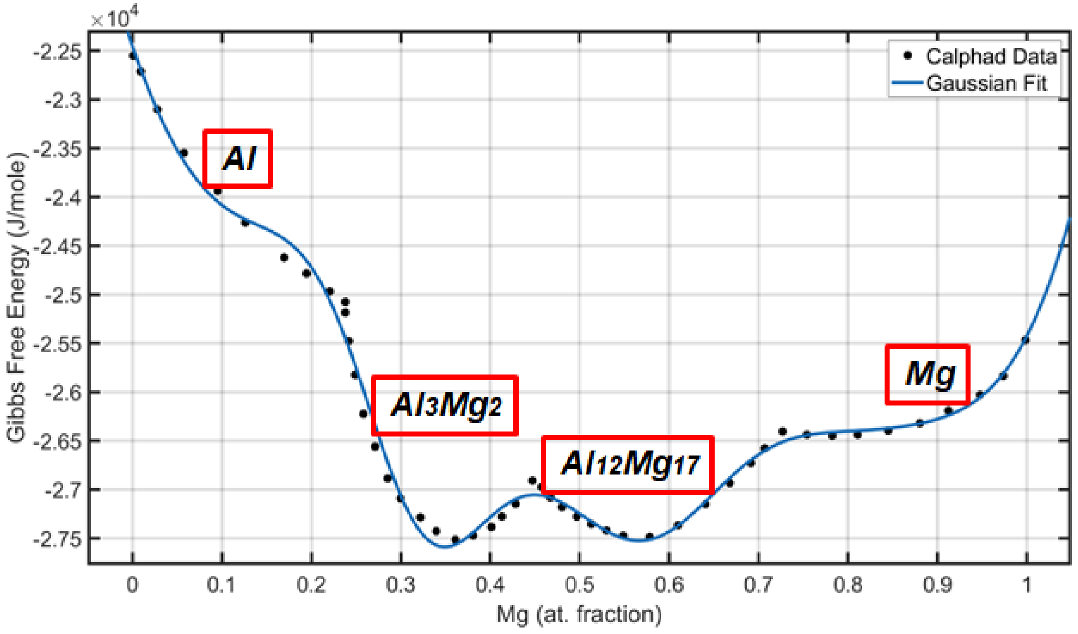


- 6022/Al_3Mg_2 interface: low Al signal and Fe rich IMC & nanoscale Mg_2Si (red box)

Diffusion Bond Modeling: Al-Mg System

Mg-Al System Gibbs Free Energy, 400°C





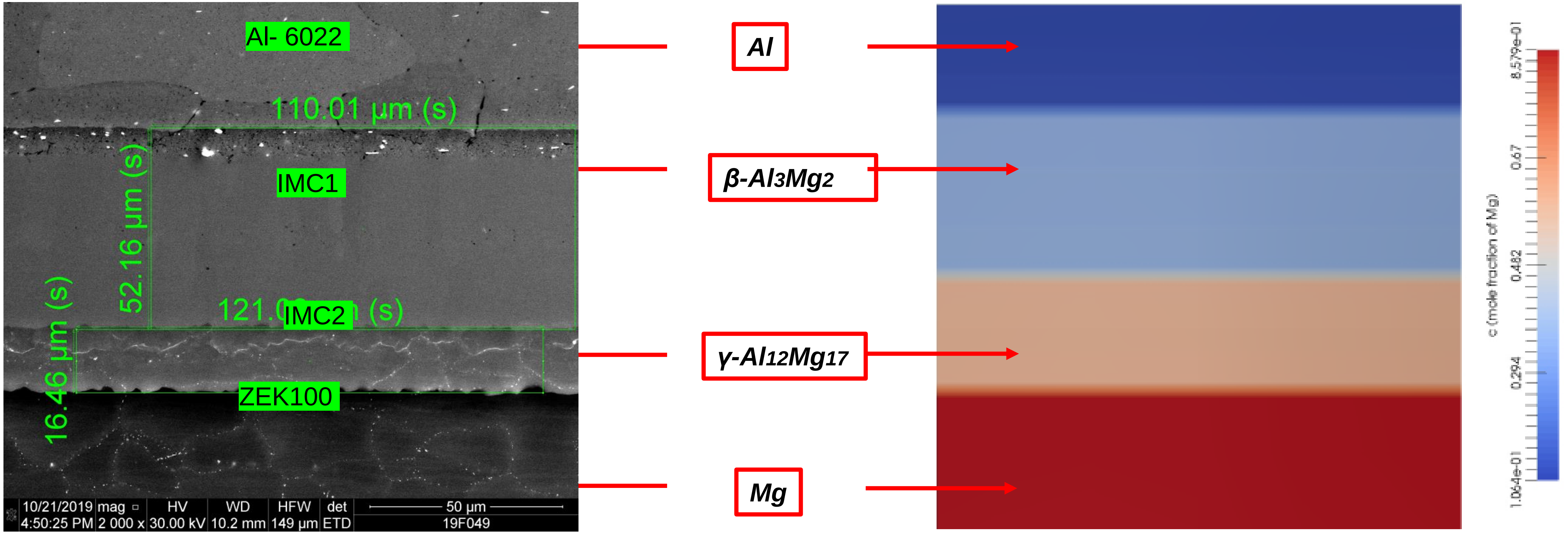
Source: Mezbahul-Islam, Mostafa, Medraj *J Matls* 2014

CALPHAD data source: ThermoCalc

Diffusion Bond Model Results

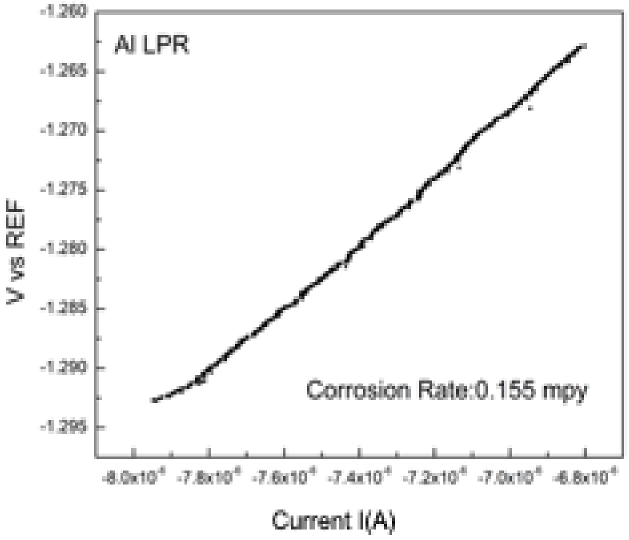
Uniform mobility → thicker β-Al_3Mg_2 layer than γ-$Al_{12}Mg_{17}$

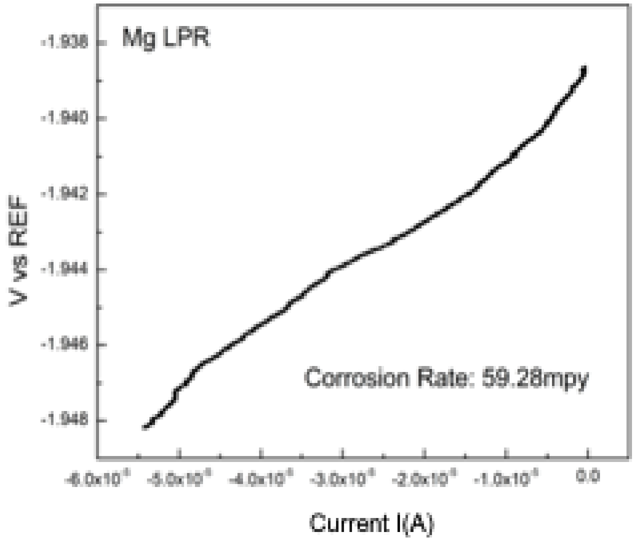
Experiment | **Phase Field Model**

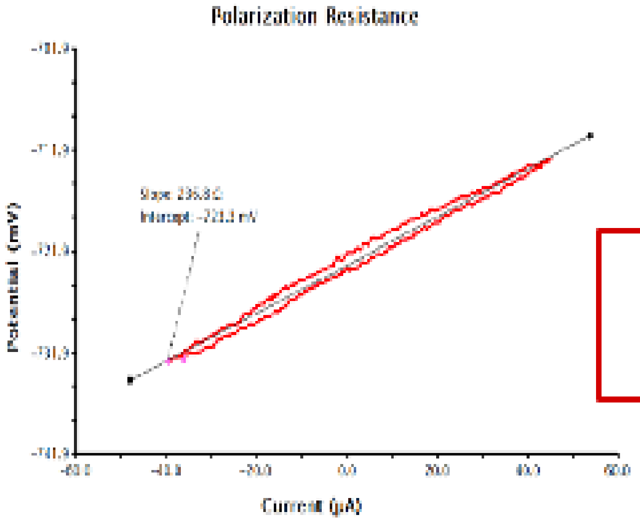


Baseline Corrosion Experiments

Linear Polarization Resistance





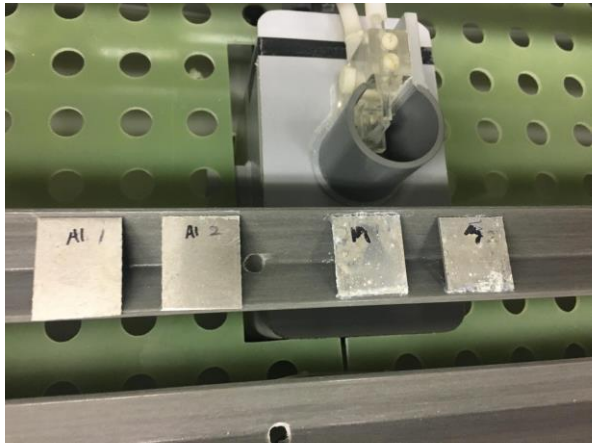


$$i_{corr} = \frac{\beta_a \beta_c}{2.3 \cdot R_p(\beta_a + \beta_c)}$$

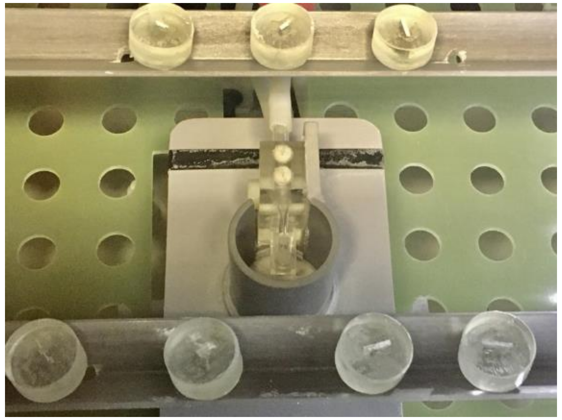
Measured corrosion rates of base metals under linear polarization agree well with literature

Cyclic Corrosion Testing (CCT)

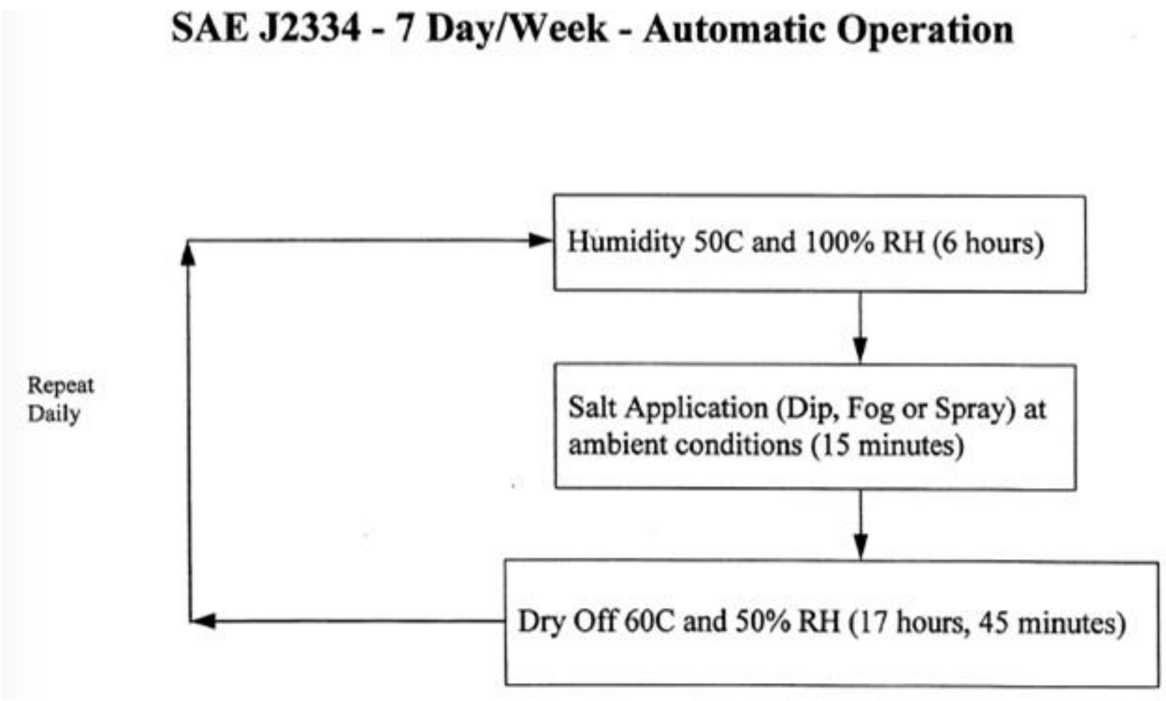
Cyclic Corrosion Test Chamber



Pure Al and Mg



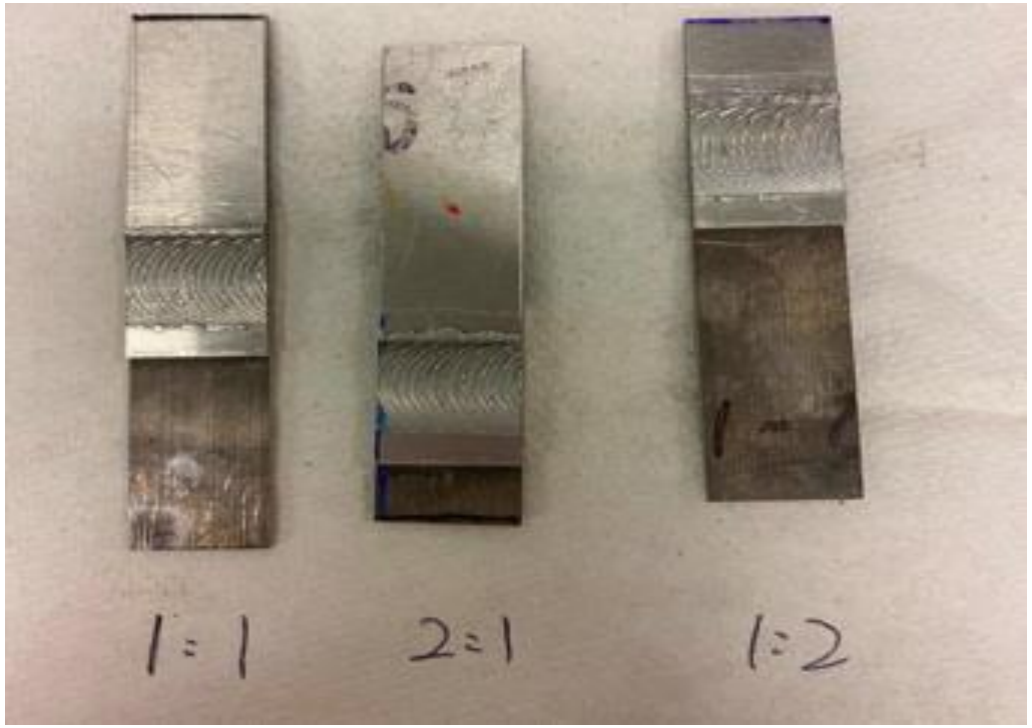
Mounted Al-Mg diffusion bonded samples



Solution: 0.5%NaCl, 0.1%CaCl2, 0.075%NaHCO3;

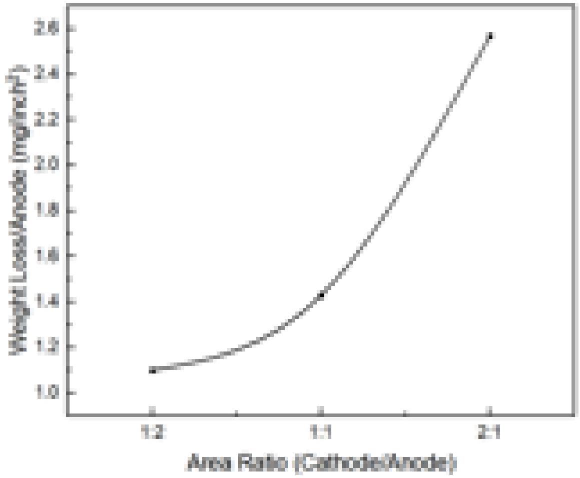
Developed and tested ability to perform cyclic corrosion tests to validate advanced corrosion model

Galvanic Corrosion Experiments



Cut samples from friction stir welds with three Cathode:anode ratios

Corrosion Rate



- 1:2 CR= 2.01 mpy
- 1:1 CR= 2.61 mpy
- 2:1 CR= 4.70 mpy

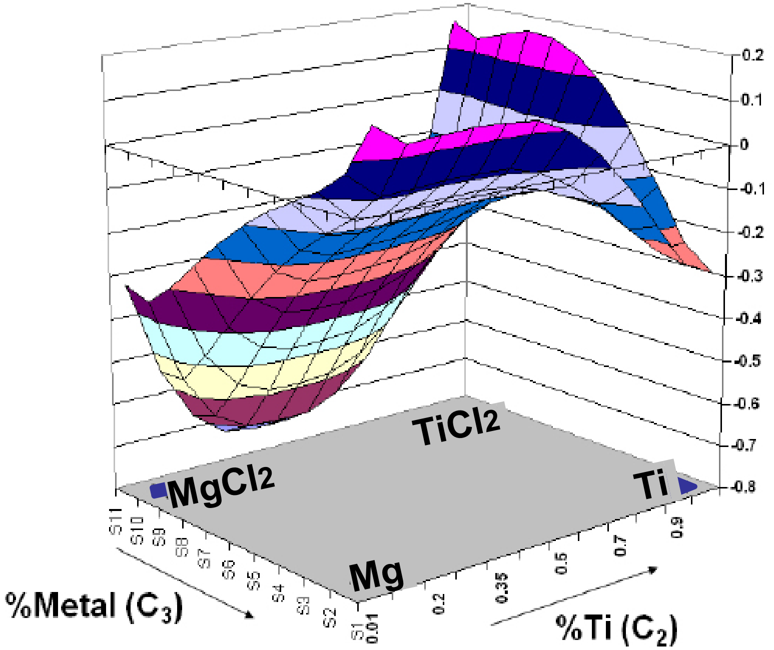
$$\text{Corrosion Rate} = \frac{534w}{DAT} \quad (\text{mpy})$$

Corrosion rate dependence on Al:Mg surface area ratio shows cathode-limited behavior in line with expectations

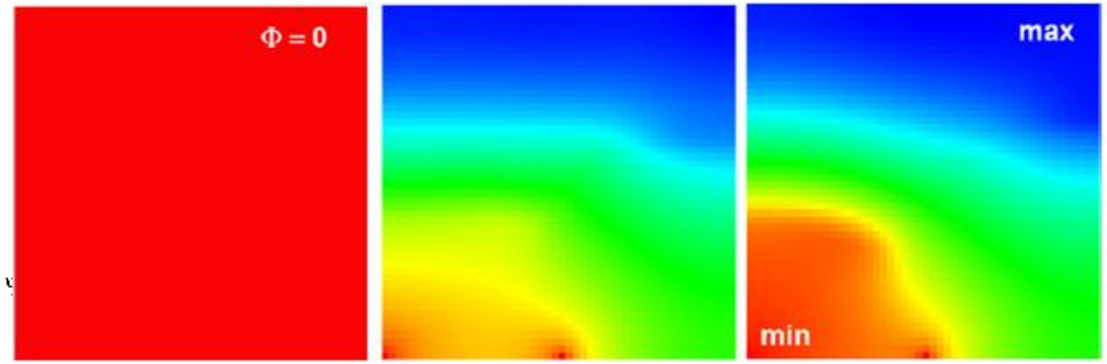
Corrosion Model Verification

- *Galvanic Reaction in Ti-Mg-Cl System*

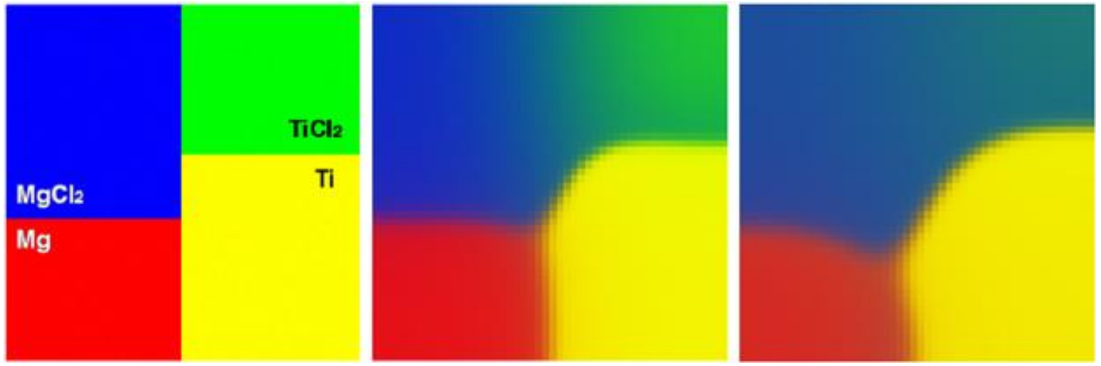
$TiCl_2$+Mg → Ti+$MgCl_2$
Reactants **Products**



Pongsaksawad *et al.* 2007

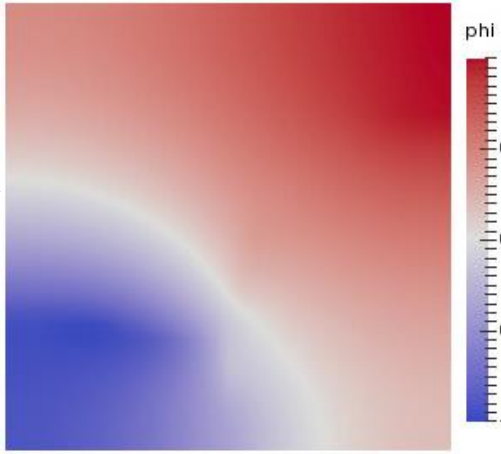


Electrical Potential

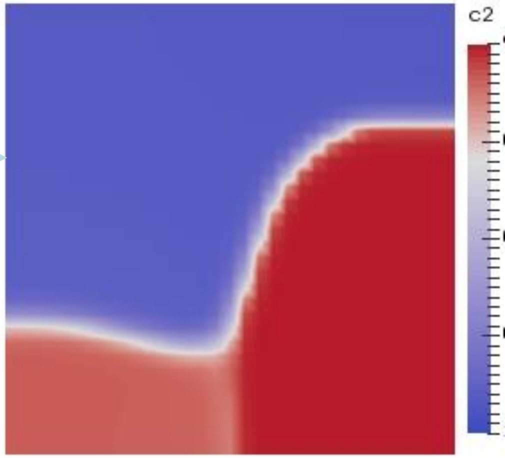


Composition

This work



Electrical Potential

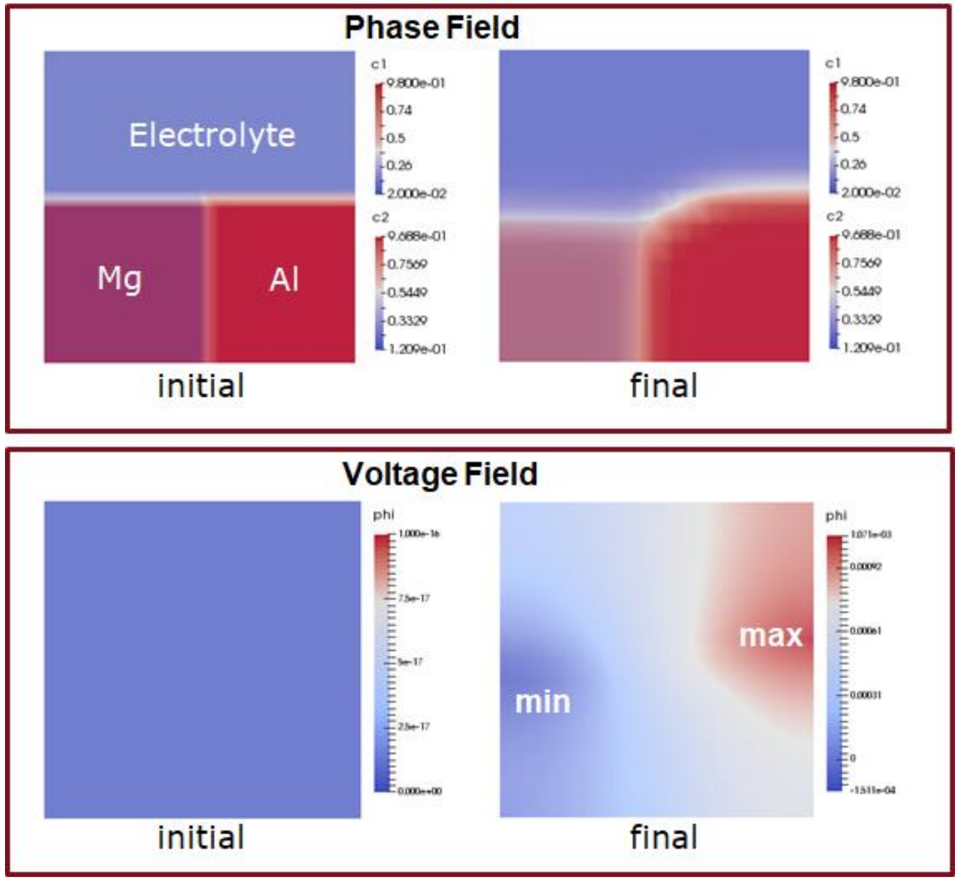


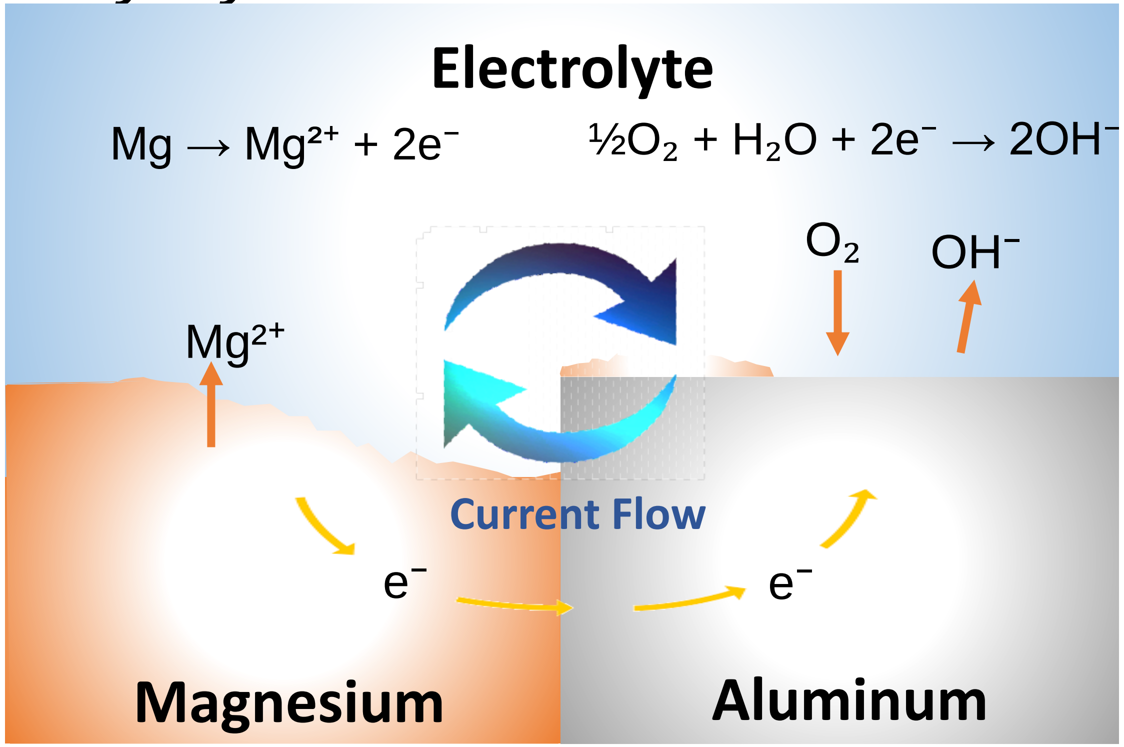
Composition

Phase field: free energy and kinetics → microstructure evolution
This formulation: charge conservation as well as free energy & kinetics
Move interfaces, change topologies automatically

Corrosion Model Preliminary Results

- *Galvanic Corrosion in Al-Mg-H2O Ternary System*





Anode (Less Noble Metal) Cathode (More Noble Metal)

Schematic of Galvanic Corrosion in Dissimilar Alloys

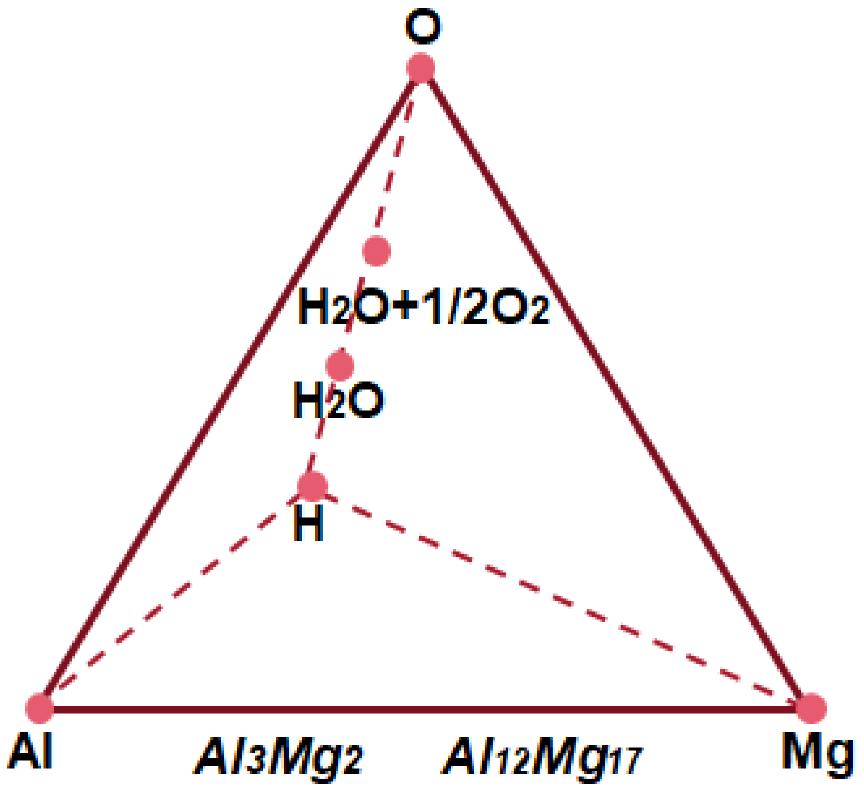
Any Al^{3+} ions in solution are reduced to Al metal at the cathode (shown here)

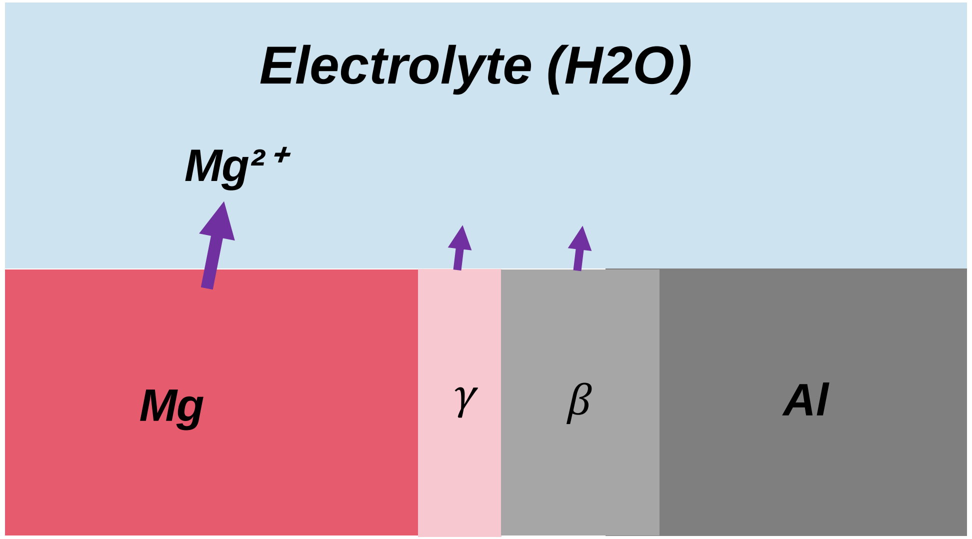
Otherwise dissolved O_2 could oxidize Al - but it's energetically favorable to oxidize Mg to Mg^{2+}

Aluminum is *cathodically protected* from oxidation by magnesium

Full Corrosion Model Formulation

Galvanic Corrosion in Al-Mg-H-O Quaternary System





Need 4-component system to address hydroxides and enable water oxidation cathode reaction: $\frac{1}{2} O_2 + H_2O + 2 e^- \rightarrow 2 OH^-$

Friction Stir Welding

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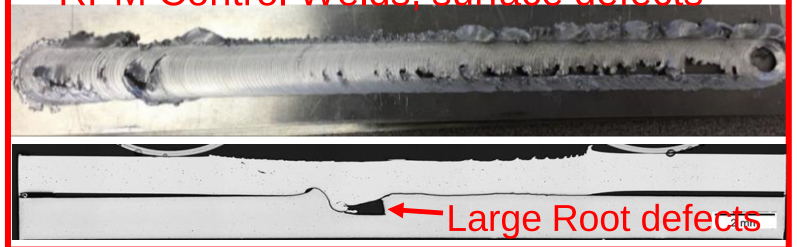
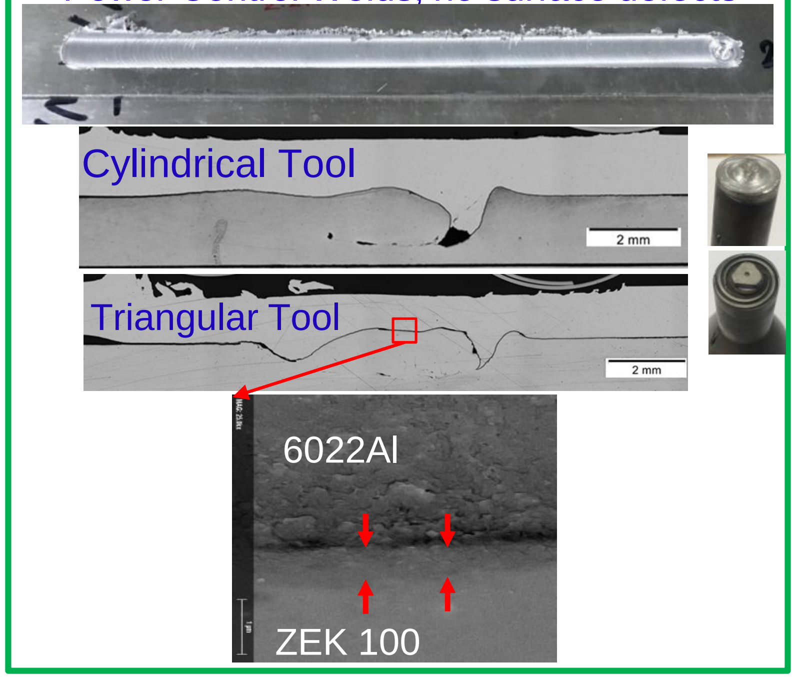
Need to weld through hard 6022 Al into soft ZEK100 Mg
Developed FSW of Mg-Al towards greater strength and repeatability. Power control method is promising.

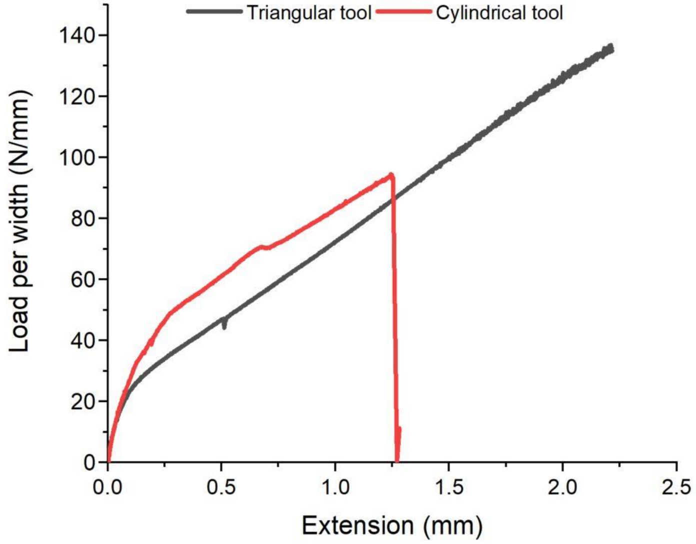
RPM Control Welds, surface defects

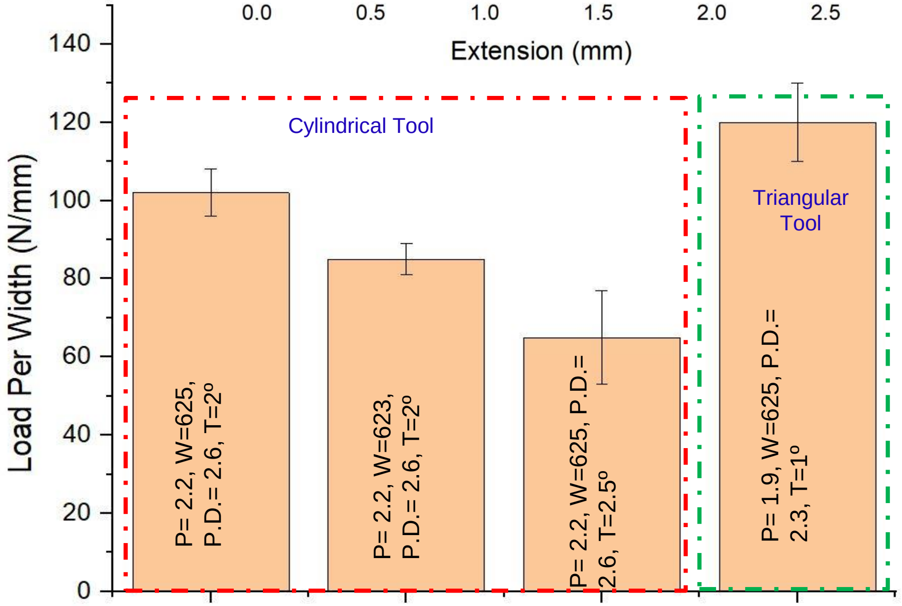


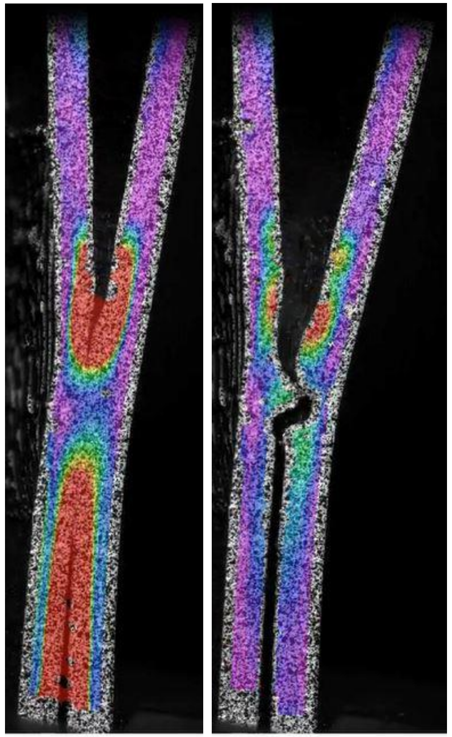
Process Development

Power Control Welds, no surface defects









Strain distribution before fracture

Response to Previous Year Reviewer Comments

Al-Mg FSW is challenging, addressing it in more detail is essential, need to plan for alternative if FSW doesn't work

- Challenge problem requirement: registration during adhesive curing
- Our goal: repeatable FSW lap strength to measure effect of corrosion
- Achieved those requirements in Budget Period 1

Validation: independent data set would forestall limited applicability

- Candidates: 7xxx Al-ZEK100 Mg, other metal pairs, fusion weld

Modeling corrosion and fracture requires electrochemistry, fracture

- Electrochemistry is described above: works for 3 components, 4-component implementation is nearly complete
- Fracture: couple with PRISMS Crystal Plasticity code

Team Collaboration & Coordination

P.I. Adam Powell of WPI leads the technical team comprised of:

WPI

Brajendra Mishra } Corrosion &
Qingli Ding } mechanics exp’ts
Kübra Karayağız – Modeling lead

PNNL

Piyush Upadhyay } Joint fabrication
Darryl Herling } & data mgt.
Erin Barker – Modeling consultant

ORNL

Donovan Leonard – Materials Characterization

Magna

Tim Skszek - Application, materials

Cost-Effective Collaboration

- Zoom - at least monthly, sometimes weekly
- One annual two-day face-to-face Project Meeting
- Team meetings at other events: TMS Annual Meeting, VTO AMR

Remaining Challenges/Barriers and Future Work

Complete corrosion model implementation and validation

- Finish galvanic corrosion tests using diffusion bonded samples
- Complete 4-component corrosion model implementation

Advanced corrosion model

- Aqueous solution corrosion → cyclic corrosion testing

Model: couple phase field corrosion with crystal plasticity mechanics

- Use FEA coupling mechanisms already in PRISMS
- Stress corrosion: need to include stress in phase field formulation?

Continue FSW development for strength and repeatability

Model cyclic loading – Erin Barker published models of FSW fatigue

Any proposed future work is subject to change based on funding levels

Summary

Validated model of Al-Mg diffusion bonding – multi-phase diffusion

Corrosion of Al, Mg, alloys, diffusion bonded couples

- Measured corrosion rates of pure Al and Mg, 6022 and ZEK100
- Verified Cahn-Hilliard phase field model of galvanic reaction
- 4-component Cahn-Hilliard formulation with water oxidation

Friction stir welding through 6022 Al into ZEK100 Mg

- Conducted extensive parametric study of FSW through Al into Mg
- Established protocol with good repeatability and strength

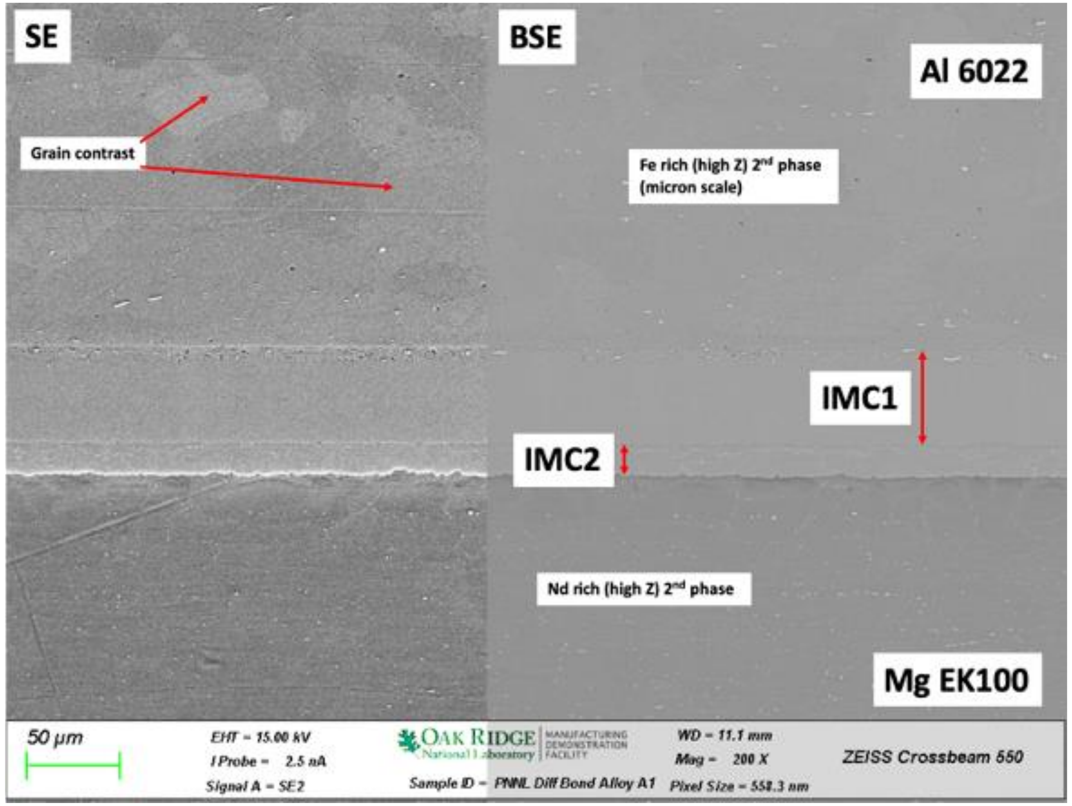
Developing the ability to model microgalvanic corrosion reactions

Potential impact on VTO objectives: Method and open source tool for modeling in a key enabler for multi-material systems

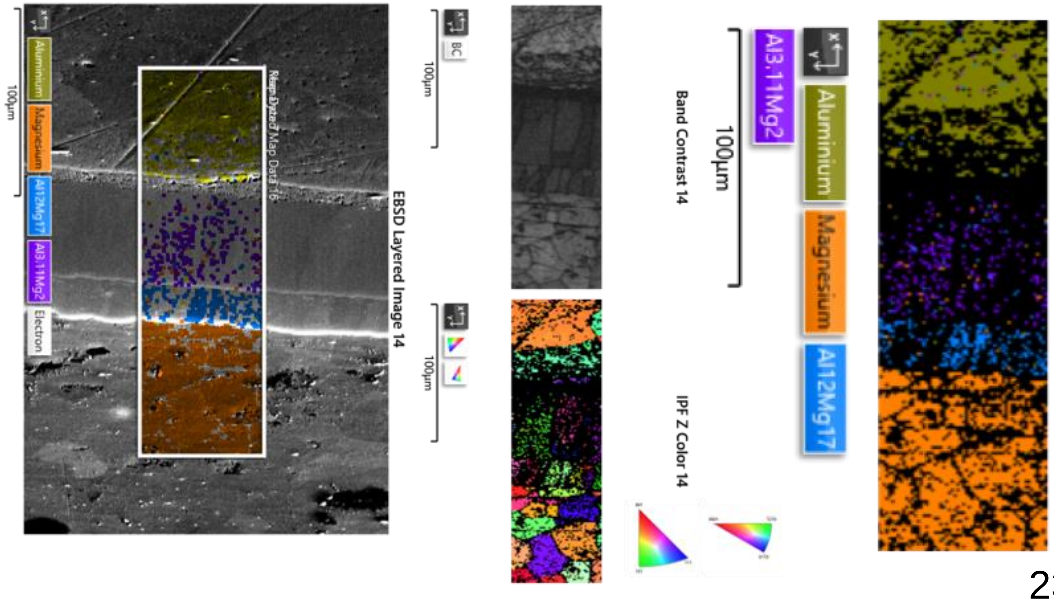
Thank You

Technical Back-Up Slides

EBSD Identification of Intermetallic Compounds in a Diffusion Bonded ZEK100/6022 Joint



- **Electron Backscattered Diffraction (EBSD) used to identify IMC1 and IMC2 (unique atomic order)**
- **Phase map (lower right) showing the 4 phases (Mg, Al, Al_3Mg_2, $Al_{12}Mg_{17}$) detected by EBSD at the diffusion bonded interface**
- **Both elemental composition and crytallagraphic orientation microanalysis protocol established**

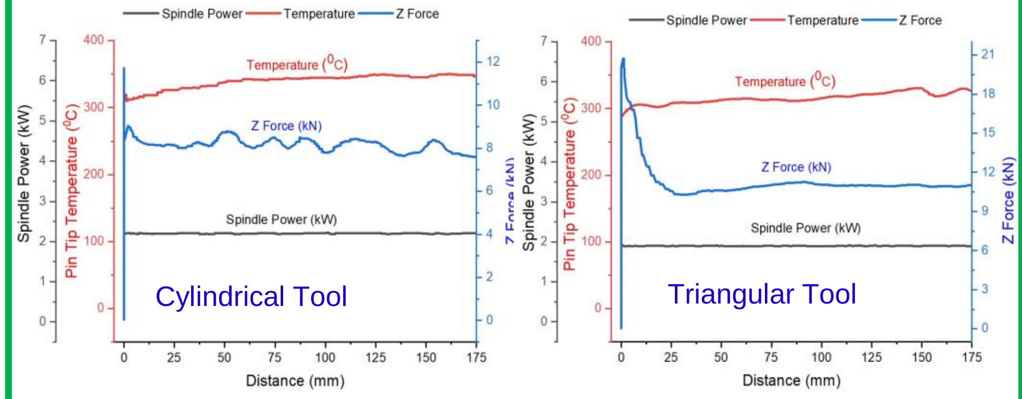


- **EDS Wt% maps: IMC1 = Al_3Mg_2 and IMC2 = $Al_{12}Mg_{17}$**
- **IMC1 ~88.5µm thick / IMC2 ~27.8µm thick**
- **3rd interfacial layer ~10µm thick at 6022/IMC1 (STEM/EDS)**

Joining Details

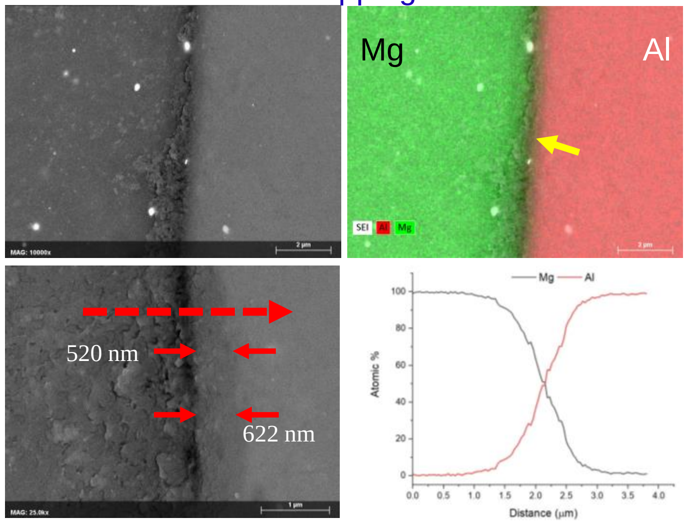
Friction Stir Welding

Power control approach

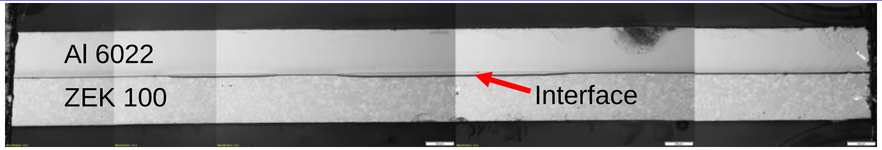


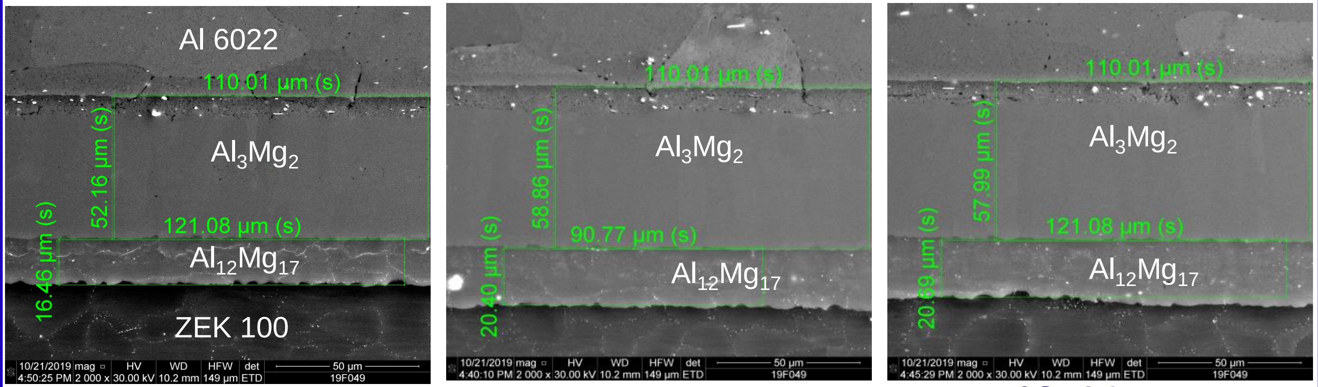
Power = 2.2 kW, .625 m/min

EDS Mapping



Diffusion Bonding





375ºC, 2 hours | 390ºC, 2 hours | 395ºC, 2 hours

Neutron Scattering (SANS) on FSW Joints: ZEK100/6022 As-Rcvd and Post-Corrosion

- **Nanoscale features of corrosion of friction stir welded aluminum and magnesium sheet metal were measured using small-angle neutron scattering (SANS) and ultra-small angle neutron scattering (USANS) instruments at the NIST Center for Neutron Research (NCNR)**

- **Preliminary SANS results are shown for stir welds corroded in 5% NaCl solutions for 0, 2, 8, and 24 hours**

- **The SANS signals increase with increasing reaction time, indicating progression in buildup of reaction products at all length scales.**

- **Data analysis and modeling will identify the composition and nanostructures of these reaction products.**